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Atomic Collisions related to Atomic Laser Isotope Separation

Takemasa SHIBATA
Department of Chemistry and Fuel Research
Japan Atomic Energy Research Institute
Tokai-mura, Ibaraki-ken, 319-11, Japan

Atomic collisions are important in various places in atomic vapor laser isotope separation (AVLIS). At a vaporization zone, many atomic collisions due to high density have influence on the atomic beam characteristics such as velocity distribution and metastable states' populations at a separation zone. In the separation zone, a symmetric charge transfer between the produced ions and the neutral atoms may degrade selectivity.

We have measured atomic excitation temperatures of atomic beams and symmetric charge transfer cross sections for gadolinium and neodymium. Gadolinium and neodymium are both lanthanides. Nevertheless, results for gadolinium and neodymium are very different. The gadolinium atom has one 5d electron and neodymium atom has no 5d electron. It is considered that the differences are due to existence of 5d electron.

Keywords: Atomic collision, Laser isotope separation, Gadolinium, Neodymium, Atomic excitation temperature, Charge transfer

1. Introduction

In atomic vapor laser isotope separation(AVLIS), one isotopic species in an atomic beam is selectively photoionized by pulse lasers and the ions are collected on electrodes. We have studied basic processes of AVLIS for gadolinium or uranium. These studies have made us recognize that atomic collisions play important roles in various places in AVLIS apparatus as shown in Fig. 1. Importance of the atomic collisions in AVLIS is summarized in Sec. 2.

Recently, we have started the atomic collision studies for neodymium. The results were expected to be similar to those for gadolinium since both gadolinium and neodymium are lanthanides and the chemical properties are very similar. However, the results for the neodymium are very different

from those for the gadolinium. Gadolinium atom has one 5d electron and neodymium atom has no 5d electron. The differences between gadoliniumm and neodymium are considered to be due to existence of 5d electron. Here it will be described that the existence of 5d electron plays important roles in collision processes in sections 3 and 4.

2. Roles of atomic collision processes in AVLIS

2.1 Collision processes in vaporization zone.

Atomic vapor is produced by heating liquid metal such as uranium or gadolinium in a water-cooled crucible with an electron beam. Since evaporated atoms are ionized by the incident and reflected electron beams, the atomic vapor contains weakly ionized plasma[1,2,3,4]. The plasma must be removed from the atomic vapor[5] before flowing into the separation zone, otherwise enrichment factor decreases.

Since atomic density is very high in the vicinity of the evaporation surface, the mean free paths of collisions between evaporated atoms are so short that the evaporated atoms have experience of many collisions which include elastic and inelastic scatterings. These collisions have influence on the atomic beam characteristics such as velocity distribution[6] and

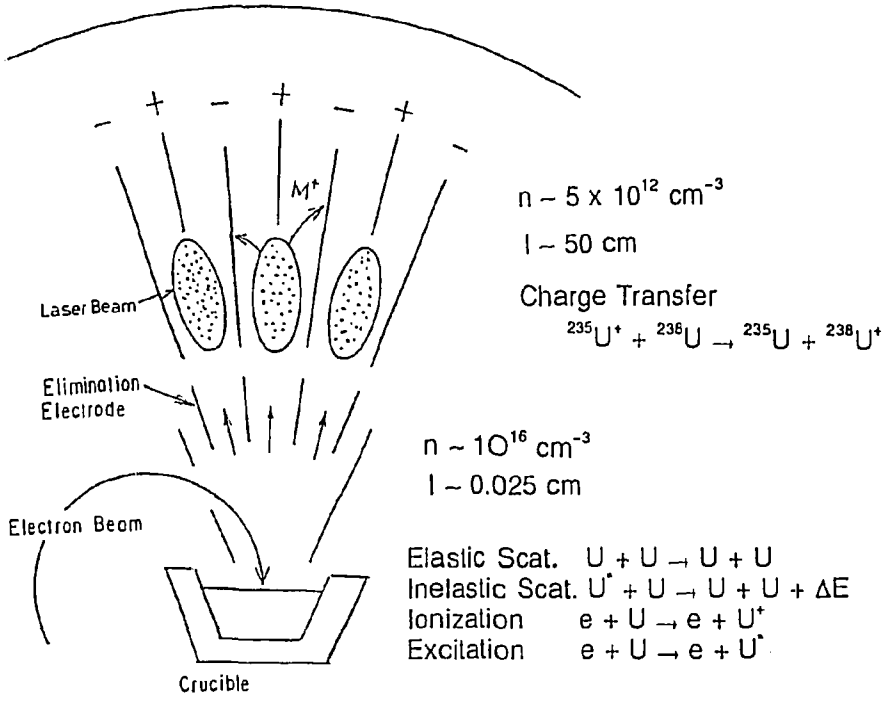

Fig.1 A schematic view of AVLIS apparatus and collision processes.

metastable states' population[7,8,9] at the laser irradiation zone. Due to vapor expansion cooling, the atomic beam velocity at the separation zone is much higher than the thermal mean velocity at the vaporization surface temperature and has a narrow distribution. The atomic excitation temperature determined from the metastable states' population is far lower than the evaporation surface temperature due to high rate energy transfer between atomic excitation and translation in the atomic vapor expanding from the evaporation surface. The measured velocity[10,11] is sometimes higher than the maximum velocity which can be attained by assuming that the total enthalpy containing the atomic excitation energy is adiabatically converted to the kinetic energy of the vapor motion. This fact suggests that the excitation energy of the atoms, which are excited the electron beams, contributes to the acceleration of the atomic vapor during expansion.

2.2 Collision processes in separation zone

In the separation zone, the atomic density is low and the mean free path is much longer than the apparatus scale such as distance between collection electrodes. Therefore, only a small part of atoms in the separation zone collides with each other. However, scattered atoms are deposited on the collection electrodes and may degrade the enrichment factor[12,13].

The symmetric charge transfer process between the produced ions and the neutral atoms degrades the enrichment factor, since the cross section of symmetric charge transfer is much bigger than that of gas kinetics. Sputtering due to impact of the photoionized ions on the collection electrodes must also be considered in designing the separation zone.

Moreover, one aimed isotope is photoionized by lasers through an autoionization level. Autoionization is a half of resonance collision of an electron with an ion. Distinct J-dependence of photoionization spectra of gadolinium atom[14] can be explained by a kind of selection rule of coupling between discrete and continuum states, where J is total angular momentum.

3. Atomic excitation temperature of gadolinium and neodymium atomic beams

The metastable states' populations in gadolinium atomic beams were measured by laser photoabsorption spectroscopy[7,8]. Moreover, metastable populations in gadolinium[9] and neodymium atomic beams were measured by the resonance photoionization. Figure 2 shows a schematic diagram of the experimental apparatus. Gadolinium or neodymium atomic beam was produced by the electron beam heating. The atoms between the collection electrodes were

ionized by two-step two-photon resonance photoionization with pulse dye lasers. The gadolinium atom has five low-lying metastable levels. Figure 3 shows resonance ionization schemes which were used to measure the population distribution of three levels of 215cm^{-1}, 533cm^{-1},and 999cm^{-1}. The first laser pulse saturated the transitions between one of the metastable states and the common intermediate state of 23644cm^{-1}. Then, the atoms in the intermediate state were ionized at the same ionization probability by the second laser pulse holding its frequency and intensity constant. Thus, the relative populations of metastable levels were obtained from the number of the photoionized ions detected by a Faraday cup. The distributions of the three levels including 0cm^{-1} or 1719cm^{-1} were also determined in the same way. The measured populations followed the Bolzmann distribution and the atomic excitation temperatures were determined. Figure 4 shows the atomic excitation temperature as a function of deposition rate monitored by a quartz crystal sensor. The excitation temperature of Gd is far lower than the evaporation surface temperature estimated from the deposition rate.

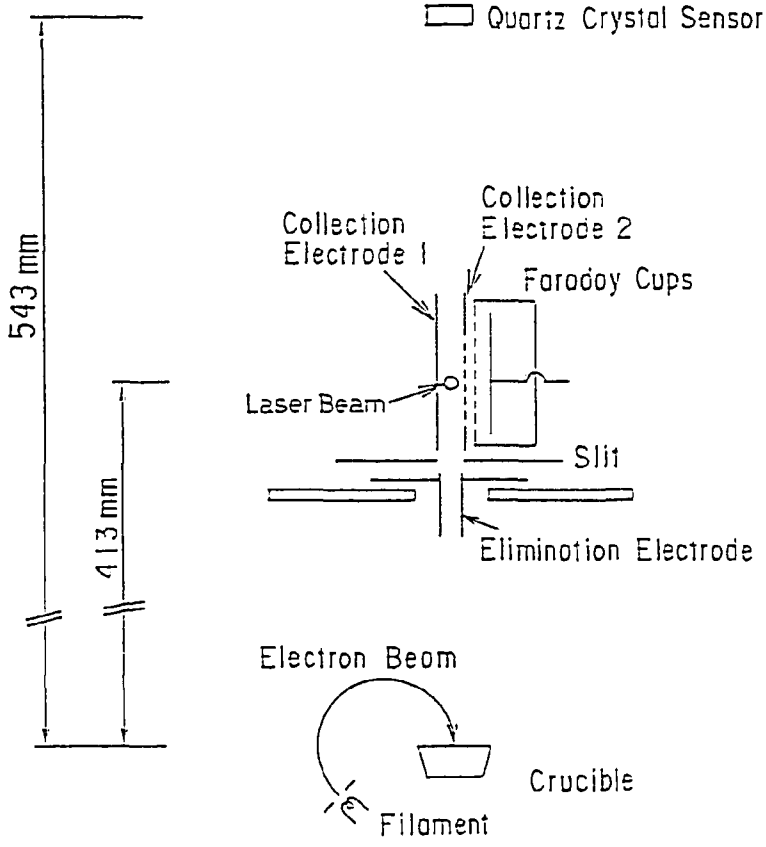

Fig.2 Schematic diagram of the experimental appratus.

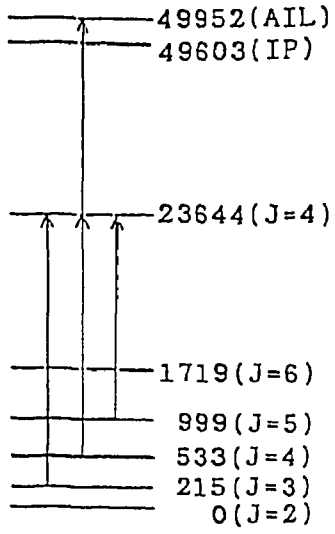

Fig.3 Ionization schemes to measure population distributions of three levels of 215, 533 and 999 cm^{-1} of gadolinium atoms.
AIL:Autoionization level
IP: Ionization potential

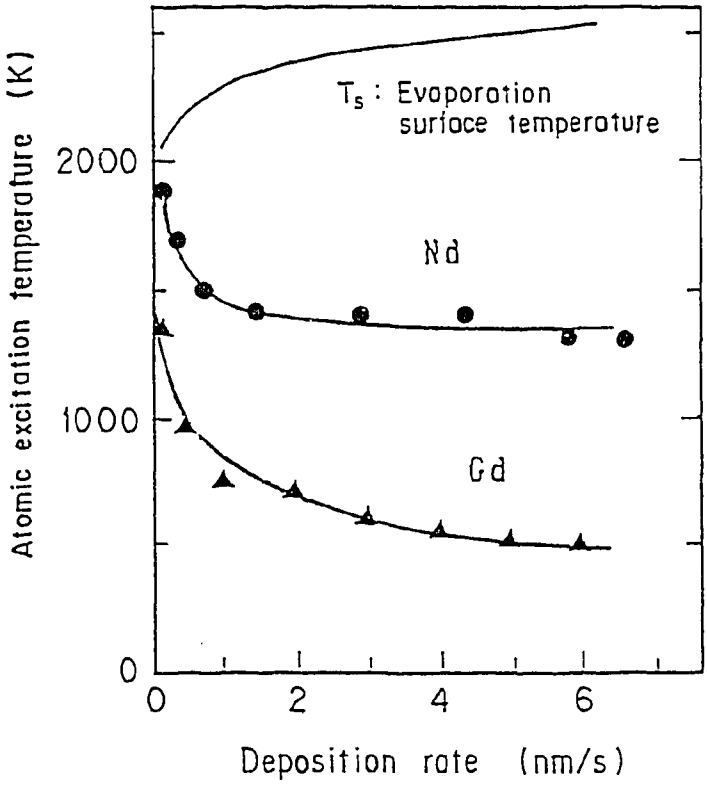

Fig.4 Atomic excitation temperatures of gadolinium and neodymium atomic beams as functions of deposition rates monitored by a quartz crystal sensor.

The neodymium atom has five low-lying metastable levels. We measured relative populations of $0cm^{-1}$ (J=4) and $1128cm^{-1}$ (J=5) using the common intermediate level of $23017cm^{-1}$ (J=4) and the autoionization level of 48533 cm^{-1}. The excitation temperature of a Nd atomic beam obtained from the relative populations of $0cm^{-1}$ and 1128 cm^{-1} is also shown in Fig. 4. The evaporation surface temperature and evaporation rate were almost similar to those for Gd evaporation. However, the atomic excitation temperature of Nd was about 1400 K at high deposition rate, which is about three times higher than temperature of Gd.

The gadolinium atom has ten valence electrons of $4f^7 5d6s^2$. The neodymium atom has six valence electrons of $4f^4 6s^2$. The collisional relaxation of gadolinium or neodymium occurs through inelastic collisions between fine structures, where the direction of spin and orbital angular momentum change. For gadolinium the direction of 6d electron easily changes by collisions and collisional relaxation easily occurs since 6d electron is slightly shielded by two 6s electrons. On the other hand, the neodymium atom has no 6d electron and ^5I states are formed by four 4f electrons which are strongly shielded by two 6s-electrons, because the orbit radius of the 4f-electrons is very short. Therefore, the direction of the spin and the orbital angular momentum is hard to change during collisions and collisional relaxation does not easily occurs. Thus, high collisional relaxation of gadolinium is considered to be due to existence of 5d electron.

4. Charge transfer cross sections of gadolinium and neodymium

Since AVLIS was proposed about 20 years ago, it has been recognized that symmetric charge transfer is an important process. However, there are few experimental data for atoms of transition elements. We measured the cross sections of gadolinium[15] and neodymium[16] from the ratio between the number of product ions and that of primary ions extracted from a laser ion source. The experimental procedures are described in detail elsewhere[15,16].

Figure 5 shows obtained cross sections of gadolinium and neodymium. The theoretical prediction of charge transfer cross section of uranium by Sinha and Bardsley[17] is also shown. The measured cross section of gadolinium roughly agrees with theoretical prediction for uranium of which ionization potential is almost the same as gadolinium. However, the cross section of neodymium is about two times of that of Gd and impact energy dependence is also different.

Usually symmetric charge transfer is resonant. However, for gadolinium or neodymium, not only resonant but also near-resonant charge transfer may

take place, since atoms or ions of gadolinium or neodymium have low-lying metastable levels. For the charge transfer between $Gd(4f^7 5d6s^2)$ and $Gd^+(4f^7 5d6s)$ which are in the ground states, two following reaction paths may take place depending on the state of spin of transferring 6s electron;

$$Gd(^9D_2) + Gd^+(^{10}D_{5/2}) \rightarrow Gd^+(^{10}D_{5/2}) + Gd(^9D_2) + 0cm^{-1}$$
$$\rightarrow Gd^+(^9D_{2/2}) + Gd(^9D_2) - 2857cm^{-1}$$

The first and second reaction paths are resonant and near-resonant, respectively. For $Nd(4f^4 6s^2)$ and $Nd^+(4f^4 6s)$ in the ground states, following resonant and near-resonant reaction paths may also take place:

$$Nd(^5I_4) + Nd^+((^5I_4)6s, J=7/2) \rightarrow Nd^+((^5I_4)6s, J=7/2) + Nd(^5I_4) + 0\ cm^{-1}$$
$$\rightarrow Nd^+((^5I_4)6s, J=9/2) + Nd(^5I_4) - 513\ cm^{-1}$$

For gadolinium, near-resonant path does not occur in the ion impact energy range of 100-1000 eV, because the energy difference is about 0.3 eV. For neodymium, on the other hand, energy difference in near-resonant reaction path is only 0.06eV. Therefore, it is considered that this near-resonant reaction occurs in the impact energy region of measurement and charge transfer cross section of Nd is bigger than that of Gd.

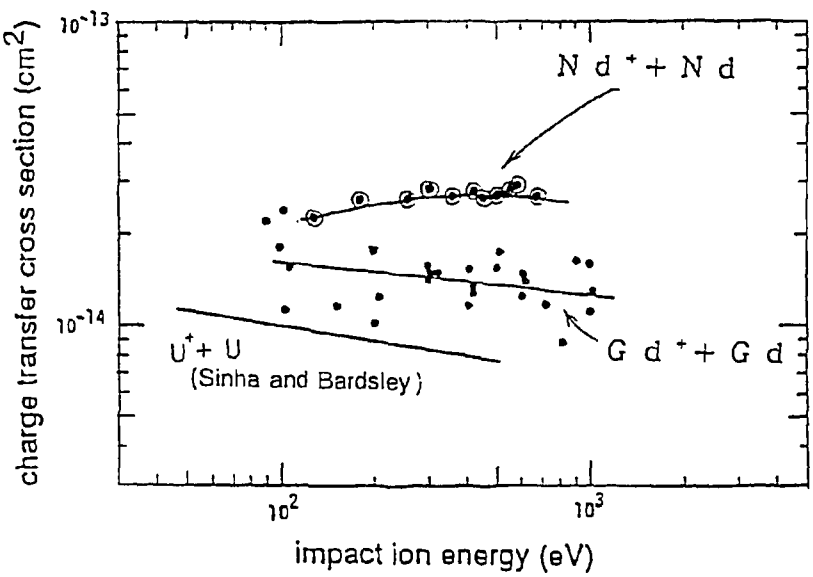

Fig.5 Measured cross sections for symmetrical charge transfer for gadolinium and neodymium as a function of the impact energy. The theoretical prediction of charge transfer cross section for uranium by Sinha and Bardsley[20] is also shown.

The Gd^+ and Nd^+ ion beam used for cross section measurement were produced by 2-step-1-wavelength resonance photoionization. From used photoionization schemes, produced Gd^+ and Nd^+ is considered to be 2857 cm^{-1} ($^{10}D_{3/2}$) and 1470 cm^{-1} (($^5I_5$)6s,J=9/2) or 1650cm^{-1} (($^5I_5$)6s,J=11/2), respectively. The atomic beams used for measurement contain low-lying metastable states as described in a previous section. Here, all possible collision paths of gadolinium and neodymium charge transfer were considered. Each cross section of possible reaction paths was calculated using the universal curve proposed by Olson[18]. Figure 6 shows total effective cross sections obtained by summing up the products of the cross section and the probability for each reaction path. As expected, the cross section of neodymium is bigger than that of Gd and energy dependence are very similar to experimental ones shown in Fig. 5, though calculated values are about 1.5 times smaller than the measured values. The energy difference between the Gd^+ levels of $^{10}D_{5/2}$ and $^9D_{3/2}$ is determined by the coupling between 5d-electron and 6s electron of Gd^+. On the other hand, the energy difference between the Nd^+ levels of (($^5I_4$)6s,J=7/2) and (($^5I_4$)6s,J=9/2) is only 0.06 eV, since Nd^+ has no 5d electron and the coupling between $4f^4$-electrons and 6s electron is small. This small energy difference is the reason why charge transfer cross section of neodymium is bigger. Therefore, the existence of d-electron is important in the case of charge transfer, also.

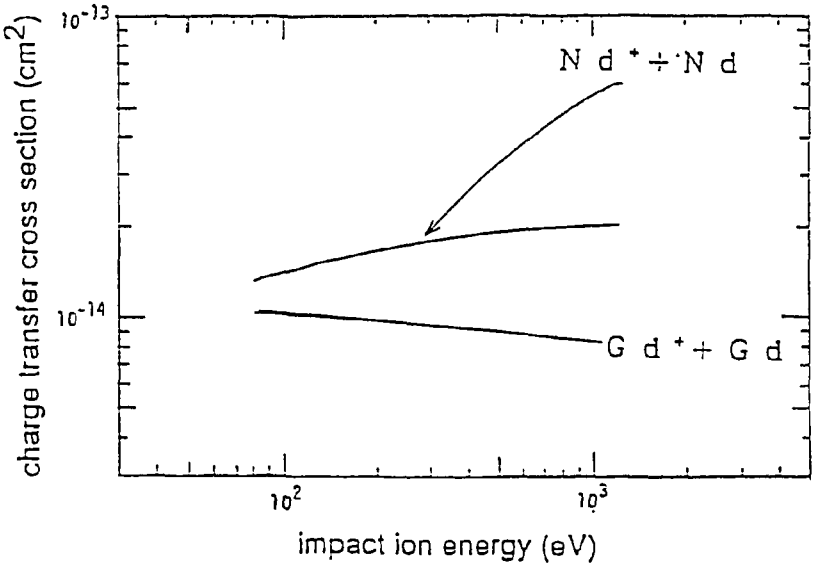

Fig.6 Calculated total effective cross sections for symmetrical charge transfer for gadolinium and neodymium.

4. Conclusion

It is briefly reviewed that atomic collisions are important in various places in AVLIS.

We have measured atomic excitation temperature of atomic beams and charge transfer cross sections of gadolinium and neodymium. The atomic excitation temperature of the gadolinium atomic beam was 500 K, which is far lower than that of the evaporation surface. On the other hand, the atomic excitation temperature of neodymium atomic beam was about 1400 K, though the evaporation surface temperature was almost similar to that for gadolinium evaporation. Symmetric charge transfer cross sections for gadolinium and neodymium were obtained in the impact energy range of 100-1000 eV. The cross section of neodymium is about two times of that of gadolinium and impact energy dependence is also different. The gadolinium atom has one 5d electron and the neodymium atom has no 5d electron. These differences between gadolinium and neodymium are explained also due to existence of 5d electron in the gadolinium atom or ion.

Uranium atom has one 6d electron in six valence electrons of $5f^3 6d 7s^2$. therefore, we can expect that the atomic collisions of uranium have the properties similar to those of gadolinium as for the atomic excitation temperature of the atomic beam and the symmetric charge transfer.

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